8th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 19–23, 2000

Semiconductor quantum dot superlattices for thermoelectric applications

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Abstract. We have theoretically investigated the in-plane lattice thermal conductivity of a quantum dot superlattice that consists of arrays of *ordered* quantum dots. Our theoretical model takes into account scattering of acoustic phonons on quantum dots made out of material with a finite acoustic mismatch with the host material. Modification of phonon dispersion in the quantum dot superlattice has also been incorporated into the model. Numerical simulation has been carried out for the quantum dot superlattice made out of multiple Si layers with Ge quantum dots. Obtained results are important for suggested applications of Si/Ge quantum dot superlattices for high-temperature thermoelectric devices.

Introduction

Quantum dot superlattices (QDS) are attractive structures that can be used in a wide range of applications [1–2]. Recently it was shown that QDS made out of suitable constituent materials might have a great potential for thermoelectric applications [3–4]. As a result, thermal properties of QDS became extremely important subject of theoretical and experimental research. Decrease of the lattice thermal conductivity due to additional phonon scattering on quantum dots and quasi-0D confinement of carriers may lead to an increase of the thermoelectric figure of merit $ZT = S^2\varepsilon/(\kappa + \kappa_e)$ (where S is the Seebeck coefficient, ε is the electric conductivity, κ is the phonon thermal conductivity, and κ_e is the electronic thermal conductivity). Acoustic impedance mismatch between material of a quantum dot and the host material (wetting layer and spacer) leads to strong phonon scattering. At the same time, electron transport is not severely deteriorated since the band-gap offset could be small (like in Si/Ge system) and the wetting layers may act as a 2D conducting channels. In this case, QDS may represent a good example of the "phonon-blocking electron-transmitting" structure with a great promise for thermoelectric applications [5].

Significant achievements in fabrication techniques for arrays of semiconductor quantum dots made possible to control both the size and position of quantum dots [6–7]. Since the size of the quantum dots and distance between two adjacent dots are comparable with the acoustic phonon wavelength, the long-range ordering of the quantum dots may produce *coherent* phonon scattering that significantly enhances phonon relaxation rates and modifies phonon group velocity. The numerical calculations were carried out for a structure that consisted of multiple layers of Si with ordered Ge quantum dots separated by wetting layers and spacers.

1. Model

The expression for the lattice thermal conductivity in the relaxation-time approximation can be written as [8]

$$\kappa = \frac{1}{3}\sum_i \mathrm{d}k v_{g_i}^2(k)\tau_{C_i} S_i(k) \tag{1}$$

where i denotes particular phonon polarization branch, v_{g_i} is the phonon group velocity, $S_i(k)dk$ is the contribution to the specific heat from modes of the polarization branch i which the phonon wave vector interval of kdk. Combined relaxation time τ_C includes all relaxation rates corresponding to the different scattering processes which do not conserve crystal momentum [9]

$$\frac{1}{\tau_C} = \sum \frac{1}{\tau} = \frac{1}{\tau_M} + \frac{1}{\tau_B} + \frac{1}{\tau_U} + \frac{1}{\tau_D}. \tag{2}$$

In Eq. (2) we included relaxation rates for three scattering processes which are dominant in Si, Ge, and $Si_\delta Ge_{1-\delta}$. These processes are $1/\tau_U$ — three-phonon Umklapp processes, $1/\tau_M$ — point defect scattering (isotopes, impurities, etc.), $1/\tau_B$ — boundary scattering, and $1/\tau_D$ — scattering on quantum dots.

The first three terms in Eq. (2) are well known [9]. The only new term we introduced the scattering on quantum dots. To describe phonon transport in QDS, we use the continuum model approximation and an assumption that the thermal phonon wave can be represented by a sum of plane waves. A phonon wave outside the dot is a superposition of incident plane and scattered waves.

In order to find $1/\tau_D$ we integrated all scattered waves from all N dots. In some arbitrary point reflected amplitude normalized to incident one Γ is

$$\Gamma = \frac{|F(\theta)|^2}{r^2}\sum_{n=1}^{N} \exp^{(iur_n)} \tag{3}$$

where $F(\theta)$ is the scattering function

$$F(\theta) = \frac{i}{2k}\sum (2n+1)(1+R_n)P_n(\cos\theta) \tag{4}$$

R_n is the dot reflection coefficient, $P_n(\cos\theta)$ are Legandre polynominals, $u = k_0 - k$, k and k_0 are the wave vectors of the plane and scattered waves, r is the radius vector.

Due to scattering on quantum dots, the phonon dispersion and corresponding phonon group velocity in QDS will be changed. This change can be taken into account using the following relation [10]

$$k'^2 = k^2\left(\left[1 + \frac{2\pi N F(0)}{Vk^2}\right]^2 - \left[\frac{2\pi N F(\pi)}{Vk^2}\right]^2\right) \tag{5}$$

where k' is the modified wave number in the presence of quantum dots, $F(0)$ and $F(\pi)$ correspond to the forward and backward scattering respectively.

Additionally, modification of the phonon group velocity may come from spatial confinement of phonon modes inside the two-dimensional spacer and wetting layers of QDS [11].

2. Results

Numerical calculations have been carried out for SiGe QDS with the spacer layer of 100 nm at temperature 300 K for different dot sizes. Lattice thermal conductivity was calculated as a function of a quantum dot volume fraction. Results are presented in Fig. 1: A, B, C curves correspond to the dot size of 1.0, 0.8, and 0.2 nm, respectively. D curve (dashed line) corresponds to the mass-different limit where each dot is just an impurity atom. Obtained results may be used for quantitative evaluation of minimum possible QDS's lattice thermal conductivity. It was found that additional decrease (about 30% at 0.1 quantum dot volume fraction) of the Si/Ge QDS can be achieved by using ordered quantum dot array instead of QDS with randomly distributed quantum dots. The additional decrease is caused by the coherent acoustic phonons scattering. The decrease of the lattice thermal conductivity remains significant in a wide temperature range.

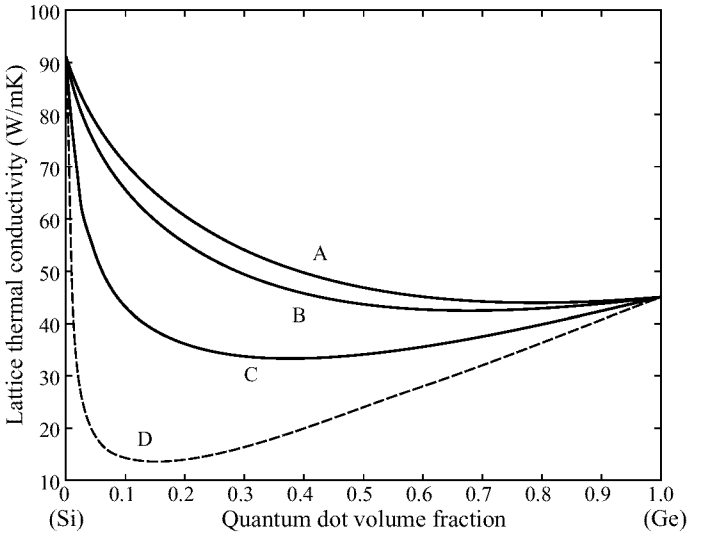

Fig. 1. Results of calculations of the lattice thermal conductivity of a quantum dot superlattice as a function of quantum dot volume fraction for different Dot sizes (a) (A—a = 1.0 nm; B—a = 0.8 nm; C—a = 0.2 nm; D—mass-different limit).

Acknowledgements

The work in UCLA was supported by the DoD MURI-ONR program on Thermoelectrics (Dr. John Pazik). The work in UCR was supported by the SRC, and the UCR seed money grant on Nanotechnology.

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